Multielectron ionization of atoms in strong fields: Classical analysis in symmetric subspace

Krzysztof Sacha[1,2] and Bruno Eckhardt[1]
[1] *Fachbereich Physik, Philipps Universität Marburg, D-35032 Marburg, Germany*
[2] *Instytut Fizyki im. Mariana Smoluchowskiego, Uniwersytet Jagielloński, ul. Reymonta 4, PL-30-059 Kraków, Poland*
(February 2, 2008)

We consider the final stage of multiple ionization of atoms in a strong linearly polarized laser field within a classical model. We propose that non-sequential multiple ionization is dominated by symmetric escape from a highly excited intermediate complex. For a configuration of N electrons with C_{Nv} symmetry in a plane perpendicular to the electric field we analyze the classical motion in phase space and discuss the final momentum distribution parallel and perpendicular to the polarization axis. The results are in good agreement with recent experiment of multiple ionization in Ne.

Double (or multiple) ionization of atoms is a fundamental process to our understanding of many electron dynamics in external fields. The surprisingly large yields of multiply charged ions reported in the first experiments with intense laser fields [1] clearly show that sequential ionization is not the leading process and that the electron-electron correlated dynamics has to be taken into account. One possibility is that the electron is shaken-off due to a non-adiabatic change in potential during the ionization of the first electron [2], a process that accounts for double ionization at high photon energies (above 1 keV) [3]. For multiphoton ionization a two step process is more likely: one electron is ionized first, accelerated by the field and driven back to the atom where it ionizes the second electron in a rescattering collision [4–10]. The applicability of tunneling expressions as in [4,11] then is due to the first ionization taking place through a tunneling process in a quasi-static electric field. Such a sequence of events is supported also by the numerical calculations of Becker and Faisal [10].

The correlation between the electrons was convincingly demonstrated in a series of recent experiments. Measurements of the distributions of ion and electron momenta in double and triple ionization [12–15] clearly show a preference for electrons escaping towards the same side of the atomic core along the field polarization axis [14]. On first sight this seems to be very different from the symmetric escape of electrons in double ionization in the absence of a field, where according to Wanniers analysis the electrons escape in opposite directions [16,17]. A closer analysis of Wanniers arguments shows, however, that they can be applied to this situation as well [18]. It is our aim here to generalize this model to the correlated escape of three or more electrons.

The essential elements of Wanniers analysis are a division of the process into two steps, the formation of a highly excited complex of two electrons close to the core and the two electron escape from this complex. At the threshold for ionization, energy is scarce. Therefore, in the dominant channel, mutual repulsion should be minimal and the energy should be equally shared by the outgoing electrons. Any deviation from the symmetric arrangement in phase space or in energy would be amplified and lead to single rather than multiple ionization. These requirements become less stringent the higher the energy above threshold.

One can argue that a similar division of the ionization process is possible in electric fields [18]. In particular, ionization also proceeds through the formation of an intermediate highly excited complex, created during the rescattering event. But the next step is different, the configuration that dominates the ionization channel is no longer the Wannier state and the threshold energy is modified as well. For instance, double ionization is observed even though the estimated energy transferred by returning electron is too small for immediate double ionization [12]. As discussed in [18] this is possible if during the collision the external field is not zero, for then a Stark saddle opens through which the electrons can escape. This saddle breaks the symmetry and focuses the electrons in the direction of the electric field. It also forces the electrons to be close to a symmetry subspace, since by mutual repulsion the electron that reaches the saddle first can push the other back to the nucleus. This would then result in either single ionization or in another rescattering event, but not in double ionization. Classical trajectory calculations within this symmetry subspace are able to reproduce the main features of the experimentally observed ion momentum distributions.

In the present letter we present a generalization of our model [18] to multielectron ionization. The key assumption is that the process is dominated by a symmetric configuration of the electrons with respect to the field polarization axis as suggested by the experiments [12–15]. Specifically, we assume that all electrons move in a plane perpendicular to the field and that they obey a C_{Nv} symmetry, which generalizes the C_{2v} symmetry of the previously analyzed case. The reflection symmetry limits the momenta to be parallel to the symmetry planes and thus confines the motion to a dynamically allowed subspace in

the high-dimensional N-body phase space. I.e. if in the full phase space of the N-body problem initial conditions are prepared in this subspace they will never leave it.

We take the electric field directed along the z-axis, and the positions of the N-electrons as $z_i = z$, $\rho_i = \rho$ and $\varphi_i = 2\pi i/N$, where (ρ_i, φ_i, z_i) are cylindrical coordinates. The momenta of the electrons are all identical, $p_{\rho,i} = p_\rho$, $p_{z,i} = p_z$ and $p_{\varphi,i} = 0$. For this geometry the classical Hamiltonian for N electrons, for zero total angular momentum along the field axis, in atomic units, with infinitely heavy nucleus and in dipole approximation, reads

$$H(p_\rho, p_z, \rho, z, t) = N\frac{p_\rho^2 + p_z^2}{2} + V(\rho, z, t), \tag{1}$$

with potential energy

$$V = -\frac{N^2}{\sqrt{\rho^2 + z^2}} + \frac{N(N-1)}{4\rho\sin(\pi/N)} + NzFf(t)\cos(\omega t + \phi) \tag{2}$$

and the pulse shape

$$f(t) = \sin^2(\pi t/T_d). \tag{3}$$

F, ω and ϕ stand for peak amplitude, frequency and phase of the external field respectively, while T_d is the pulse duration.

Experiments show that the multiple ionization is possible even when the energy transferred by the rescattered electron is less than that needed to lift the system to the many electron continuum. We therefore assume that the complex from which multiple ionization starts is described by Hamiltonian (1) with initially negative energy E. Moreover, since the electronic motion close to the nucleus is much faster than the changes in the electric field the process of electron escape can be discussed adiabatically for fixed external field. This allows us to analyze the potential energy for a fixed time.

Before we proceed with the analysis we take advantage of the scaling symmetry of the classical Hamiltonian (1) and eliminate one parameter. If the variables are rescaled according to

$$\begin{aligned} H &\to \sqrt{F}H, \\ \rho,\ z &\to \rho/\sqrt{F},\ z/\sqrt{F}, \\ p_\rho,\ p_z &\to F^{1/4}p_\rho,\ F^{1/4}p_z, \\ \omega,\ t &\to F^{3/4}\omega,\ F^{-3/4}t \end{aligned} \tag{4}$$

the dynamics becomes independent of the peak value of the field amplitude, i.e. we obtain the system described by the Hamiltonian (1) with $F = 1$.

Equipotential curves of Eq. (2) in the scaled variables (4), for $N = 3$ and $N = 6$ are shown in Fig. 1. The saddles are located along the lines $z = r_s\cos\theta_s$ and $\rho = r_s\sin\theta_s$ with $\theta_s = \theta$ or $\theta_s = \pi - \theta$ where

$$\theta = \arctan\left[\left(\frac{4N\sin(\pi/N)}{N-1}\right)^{2/3} - 1\right]^{-1/2} \tag{5}$$

and

$$r_s^2 = \frac{N}{f_t}\sqrt{1 - \left(\frac{N-1}{4N\sin(\pi/N)}\right)^{2/3}} \tag{6}$$

with $f_t = |f(t)\cos(\omega t + \phi)|$. The energy of the saddle is

$$V_s = -\sqrt{4N^3 f_t}\left[1 - \left(\frac{N-1}{4N\sin(\pi/N)}\right)^{2/3}\right]^{3/4}. \tag{7}$$

During a field cycle the saddle moves in from infinity along the line $\theta_s = \theta$, back out to infinity and then in and out again along the line $\theta_s = \pi - \theta$. The angle θ increases with the number of electrons N, while the energy of the saddle changes non-monotonically as shown in Fig. 2. For $N \geq 14$ the saddle disappears. For this many electrons the repulsion between the electrons is stronger than the attraction to the nucleus, see Eq. (2) for $z = 0$. The situation with $N \geq 14$ is rather an extreme case from the experimental point of view but it reflects the fact that a bound state for a neutral atom with N electrons symmetrically distributed in a plane can not be formed if $N \geq 14$.

The saddle which is opened by the external field allows electrons to escape even if the total energy E of the highly excited complex is negative. It is enough, classically, that E is greater than the energy of the saddle. The saddle thus provides a kind of transition state [19,20] for the ionization process: once the electrons cross it, they are accelerated by the field and pulled further away, making a return rather unlikely. Moreover, they can acquire the missing energy so that the electrons can escape even when the field vanishes. The field thus plays a double role in determining a threshold for this process: during the first stages of the rescattering process it provides the energy for the collision complex and during the final stages it opens the path for multiple escape.

In the experiment [15] of triple (also single and double) ionization of Ne by ultrashort (30 fs) laser pulses at 795 nm and at intensities 1.5 PW/cm^2 the distributions of momenta of ions parallel and perpendicular to the polarization axis have been measured. In the limit of negligibly small momentum transfer by the absorbed photons, the ion momentum $\vec{p}_{ion}$ reflects the sum of the momenta of the emitted electrons, $\vec{p}_{ion} = -\sum_i \vec{p}_i$ [12,15]. To calculate the experimental distributions corresponding to the triple ionization of Ne we have performed classical trajectory simulations for $N = 3$. Note that the correct equations of motion for a single electron follow from the Hamiltonian (1) after division by N.

The Hamiltonian (1) with $N = 3$ is the full Hamiltonian for a Li with its three electrons. In order to relate it to triple ionization from atoms with more electrons the zero point in energy has to be shifted to the threshold for

the three electron continuum, and all energies have to be taken relative to this level. Moreover, interactions with the electrons in the core are neglected. Specifically, for the modeling of the experiments [15] on triple ionization in Ne we thus assume that in a rescattering process the energy transfer is less than the threshold for triple ionization (about 4.6 a.u.). The precise value of the initial energy E depends on the details of the rescattering process and can thus not be determined within our model: it is a free parameter. The rescattering event is most likely to take place when the field amplitude is high, so we have started the simulation close to the top of the pulse, i.e. for $t_0 = 0.33T_d$, where $T_d = 2412$ a.u. corresponds to the experimental pulse duration [15]. The corresponding amplitude and frequency of the field are $F = 0.207$ a.u. and $\omega = 0.057$ a.u. For such field parameters the saddle energy equals -3.5 a.u. We have performed numerical simulations for several initial energies. The distributions of the electron momenta for $E = -1$ a.u. are shown in Fig. 3. The distribution for parallel momenta can be compared with the corresponding experimental distribution of ions momenta [15]. The main features of the distribution, its width and characteristic double hump structure, are well reproduced in our calculation. The minimum in the distribution is, however, less pronounced than that observed in the experiment. Overall, we take this good agreement as strong indication that triple ionization can only occur in the neighborhood of the symmetric process discussed here.

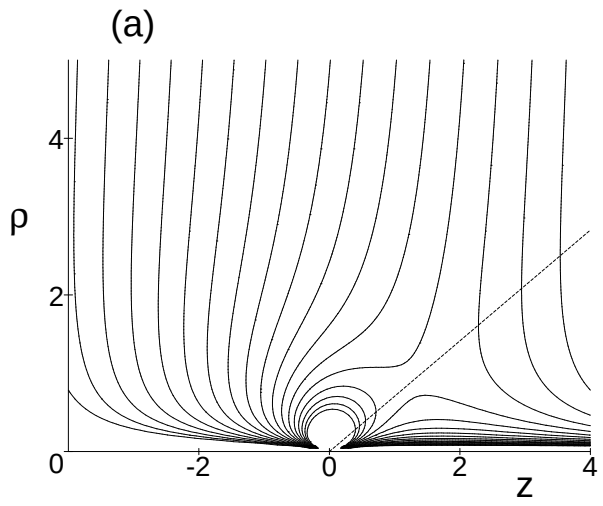
For initial energies below about -3 a.u. the double hump structure disappears and only a single maximum remains. This corresponds to experiments with weaker fields which transfers less energy to the rescattered electron. Then the electrons crossing the saddle have smaller kinetic energy and the interaction with the remainder of the pulse blurs the distribution. Such a change in the character of the distribution with decrease of field intensity has also been observed in the experiment of double ionization of He atoms [12].

The distribution for perpendicular ion momenta can not be calculated in our model since this component vanishes exactly by symmetry. But we can calculate the transverse momentum of an individual escaping electron, as shown in Fig. 3. The absence of events around zero reflects simply the effect of the repulsion between electrons. Measurements of this quantity should provide further test of this model.

In conclusion we have shown that the double humped structure in ion momenta distribution observed in the experiment of triple ionization of Ne [15] can be calculated within a classical simulation assuming symmetrically escaping electrons. This multi-ionization path can be extended to include up to 13 electrons. The successful comparison with the experimental data supports the idea that the dominant contribution to ionization comes from initial conditions in the highly excited collision complex that are asymptotic to this symmetry plane, very much as in Wanniers analysis [16,17,21].

We would like to thank Harald Giessen for stimulating our interest in this problem and for discussions of the experiments. Financial support by the Alexander von Humboldt Foundation and by KBN under project 2P302B00915 are gratefully acknowledged.

[1] A. L'Huiller, L. A. Lompre, G. Mainfray, and C. Manus, Phys. Rev. A **27**, 2503 (1983).
[2] D.N. Fittinghof, P.R. Bolton, B. Chang, and K.C. Kulander, Phys. Rev. Lett. **69**, 2642 (1992).
[3] V. Schmidt, *Electron Spectrometry of Atoms using Synchrotron Radiation* (Cambridge University Press, Cambridge, England, 1997).
[4] P.B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993)
[5] K.C. Kulander, J. Cooper, and K.J. Schafer, Phys. Rev. A **51**, 561 (1995)
[6] A. Becker and F.H.M. Faisal, J. Phys. B **29**, L197 (1996)
[7] A. Becker and F.H.M. Faisal, J. Phys. B **32**, L335 (1999)
[8] B. Sheehy, R. Lafon, M. Widmer, B. Walker, L.F. DiMauro, P.A. Agostini, and K.C. Kulander, Phys. Rev. A **58**, 3942 (1998)
[9] A. Becker and F.H.M. Faisal, Phys. Rev. A **59**, R1742 (1999)
[10] A. Becker and F.H.M. Faisal, Phys. Rev. Lett. **84**, 3546 (2000).
[11] U. Eichmann, M. Dörr, H. Maeda, W. Becker, and W. Sandner, Phys. Rev. Lett. **84**, 3550 (2000).
[12] Th. Weber, M. Weckenbrock, A. Staudte, L. Spielberger, O. Jagutzki, V. Mergel, F. Afaneh, G. Urbasch, M. Vollmer, H. Giessen and R. Dörner, Phys. Rev. Lett. **84**, 443 (2000)
[13] Th. Weber, M. Weckenbrock, A. Staudte, L. Spielberger, O. Jagutzki, V. Mergel, F. Afaneh, G. Urbasch, M. Vollmer, H. Giessen and R. Dörner, J. Phys. B: At. Mol. Opt. Phys. **33**, L1 (2000)
[14] Th. Weber, H. Giessen, M. Weckenbrock, G. Urbasch, A. Staudte, L. Spielberger, O. Jagutzki, V. Mergel, M. Vollmer, and R. Dörner, Nature **???**, ??? (2000) (in press)
[15] R. Moshammer, B. Feuerstein, W. Schmitt, A. Dorn, C.D. Schöter, J. Ullrich, H. Rottke, C. Trump, M. Wittmann, G. Korn, K. Hoffmann and W. Sandner, Phys. Rev. Lett. **84**, 447 (2000)
[16] G.H. Wannier, Phys. Rev. **90**, 817 (1953)
[17] A. R. P. Rau , Phys. Rep. **110**, 369, (1984)
[18] B. Eckhardt and K. Sacha, physics/0005079.
[19] E. P. Wigner, Z. Phys. Chemie B **19**, 203 (1932); Trans. Faraday Soc. **34**29, (1938)
[20] E. Pollak, in *Theory of Chemical Reactions*, vol III, M. Baer, ed., (CRC Press, Boca Raton, 1985, p. 123
[21] J.M. Rost, Phys. Rep. **297**, 271 (1999)



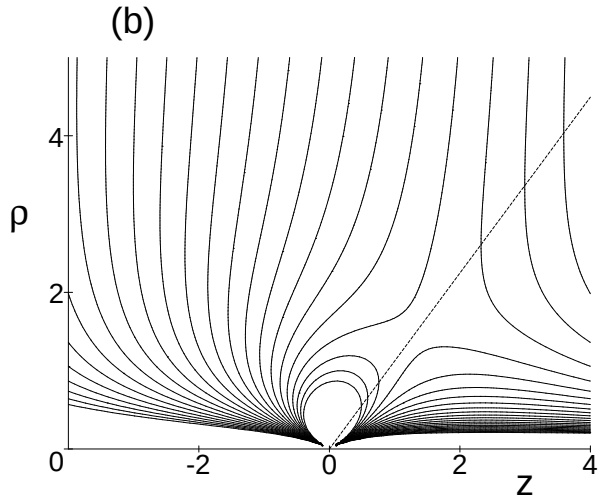


FIG. 1. Equipotential curves of the potential energy (2), in the scaled variables (4), for two different values of the number of electrons: $N = 3$ [panel (a)] and $N = 6$ [panel (b)]. The potentials are plotted for fixed time corresponding to the maximal field amplitude. The saddles move along the dashed lines when the electric field points in the positive z-direction and along the second obtained by reflections on $z = 0$ during the other half of the field cycle. Note that the angle between the z-axis and the dashed line increases with the number of electrons.

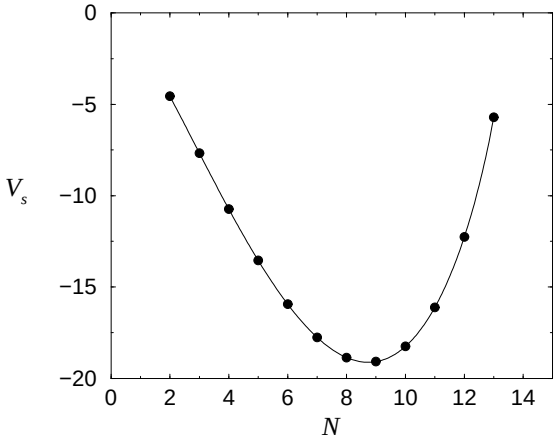


FIG. 2. The energy of the saddle point of the potential (2), in the scaled variables (4), versus number of electrons N. The values correspond to the maximal field amplitude.

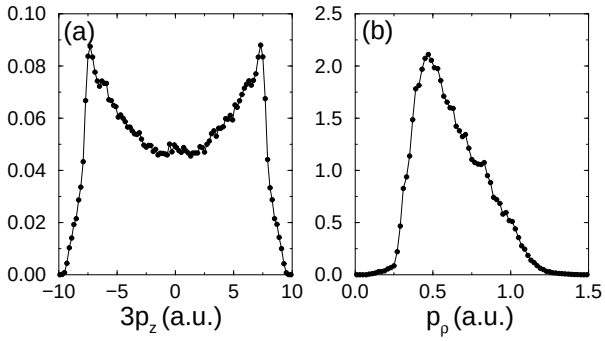


FIG. 3. Final distribution of electrons momenta parallel [panel (a)] and perpendicular [panel (b)] to the field polarization axis for $E = -1$ a.u. Panel (a) corresponds to the experimental distribution of ions momenta measured in the triple ionization of Ne [15]. Note that, in panel (b), the distribution rapidly falls to zero around $p_\rho \approx 0$ reflecting the effect of repulsion between the electrons. The distributions are based on an ensemble of $2 \cdot 10^5$ trajectories.